Single-photon exchange interaction in a semiconductor microcavity

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We consider the effective coupling of localized spins in a semiconductor quantum dot embedded in a microcavity. The lowest cavity mode and the quantum dot exciton are coupled and close in energy, forming a polariton. The fermions forming the exciton interact with localized spins via exchange. Exact diagonalization of a Hamiltonian in which photons, spins and excitons are treated quantum mechanically shows that *a single polariton* induces a sizable indirect exchange interaction between otherwise independent spins. The origin, symmetry properties and the intensity of that interaction depend both on the dot-cavity coupling and detuning. In the case of a (Cd,Mn)Te quantum dot, Mn-Mn ferromagnetic coupling mediated by a single photon survives above 1 K whereas the exciton mediated coupling survives at 15 K.

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Control of exchange interactions in solid state environments has become a strategic target in the development of both spintronics and quantum computing [1, 2, 3, 4, 5]. Artificial control of *direct* exchange interactions, which occur at length scales of one lattice spacing, is hardly possible with current day technologies. In contrast, there is a number of proposals to control *indirect* exchange interactions (IEI) of distant spins sitting several nanometers away [1, 2, 3, 6, 7, 8, 9, 10], taking advantage of the optical and electrical manipulation of the intermediate fermions afforded in semiconducting hosts. The local spins could be provided by the nuclei [3], by electrons bound to donors [6, 8], or d electrons of magnetic impurities [9, 10]. Artificial control of the IEI has been observed experimentally giving rise to a variety of phenomena, like the reversible modification of the Curie temperature and coercive fields in (III,Mn)V [11] and (II,Mn,N)VI semiconductors [12], the induction magnetic order in otherwise paramagnetic (II,Mn)VI semiconductor quantum dots [13] and the enganglement of donor spins in (II,Mn)VI quantum wells [8]. Such a control is also a must in the implementation of quantum computation using localized spins in solids, since 2 qbit operations require exchange interactions between distant spin pairs [14, 15].

Laser driven IEI in semiconductors is particularly promising because it affords control in the time domain and it can be tuned by changing the laser frequency, intensity and polarization. Above gap excitation [8, 12, 13] creates real carriers that mediate 'RKKY like' exchange. Below gap excitation induces an optical coherence between conduction and valence band able to mediate exchange interactions between localized spins[6, 10]. The strength of the 'optical RKKY' exchange interaction (ORKKY) is determined both by the intensity of the laser, proportional to the square of the Rabi energy Ω [6], and by the detuning $\delta = E_g - \omega_L$ between the semiconductor gap and the laser frequency. When $\delta > \Omega$, the laser-matter coupling can be treated in perturbation theory [6] and the strength of the coupling is proportional to Ω^2/δ^3.

Here we study the interaction between two spins located in a quantum dot embeded in a microcavity tuned close to resonance with the quantum dot semiconductor gap. The dot provides full three dimensional confinement for the electrons and the holes and the cavity provides full three dimensional confinement for the photon field. A large value of the electromagnetic density is obtained due to photon confinement in a microcavity without the use of ultra-short laser pulses [16]. The exchange between the intermediate fermions and the localized spins is also enhanced if the former are confined [9]. We find that confinement of both the light and the intermediate fermions yields an enhancement of the ORKKY interaction so big that *a single photon can induce an indirect optical exchange interaction* between two localized spins at temperatures of 1 Kelvin. Confinement also permits the *exact* diagonalization of the Hamiltonian, considering all the localized spins, the intermediate fermions and the electromagnetic field *fully quantum mechanically.* The exact solution of the problem provides an unified treatment of carrier mediated and optical IEI.

In the following, we consider a micrometer size cylindrical cavity [16], made of CdTe with inclusions of (Cd,Mn)Te quantum dots [17]. We consider small parallelepiped dots [18] that confine conduction band electrons

(creation operator $c^\dagger$) and valence band holes (creation operator $d^\dagger$) with intra-band level spacing larger than all the other intra-band energy scales in the problem, so that we only keep the lowest orbital level in each band, ϵ_c and ϵ_v. These levels have a twofold spin degeneracy. Their orbital wave functions are $\psi_e(\vec{r})$) and $\psi_h(\vec{r})$ respectively.

The electric field of the lowest cavity mode lies mainly in the plane perpendicular to the axis of the cylinder. In consequence, there are two degenerate cavity modes, associated to the two possible polarization states in that plane. Their energy $\hbar\omega_0$ is close to the quantum dot band gap, E_g. We choose the circularly polarized cavity modes as a basis and, after canonical quantization, the corresponding photon creation operator is denoted by $b^\dagger_\lambda$, where $\lambda = L, R$. The most general Hamiltonian we consider can be splitted in 4 terms, $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_g + \mathcal{H}_J + \mathcal{H}_U$. The first reads

$$\mathcal{H}_0 = \sum_\lambda \hbar\omega b^\dagger_\lambda b_\lambda + \sum_\sigma \left[\epsilon_v d^\dagger_\sigma d_\sigma + \epsilon_c c^\dagger_\sigma c_\sigma\right] \tag{1}$$

and describes the Hamiltonian for decoupled cavity photons and quantum dot fermions. The light-matter coupling only has non diagonal terms in the band index:

$$\mathcal{H}_g = \sum_{\sigma_e,\sigma_h,\lambda} G^\lambda_{\sigma_e,\sigma_h} \left(b^\dagger_\lambda + b_\lambda\right) \left[c^\dagger_{\sigma_e} d^\dagger_{\sigma_h} + d_{\sigma_h} c_{\sigma_e}\right] \tag{2}$$

If we assume that the hole is purely heavy, we obtain the standard spin selective coupling [6, 10, 19] $G^\pm_{\sigma_e,\sigma_h} = \frac{g}{2}\left(\delta_{\sigma_e,\sigma_h} \pm \hat{z}\cdot\vec{\tau}_{\sigma_e,\sigma_h}\right)$ where g is the Rabi energy and $\vec{\tau}$ are the Pauli matrices. This coupling breaks spin rotational invariance and privileges the axis of the cavity, $\hat{z}$. The value of g depends on the amplitude of the cavity mode in the location of the dot and plays the same role than the Rabi energy Ω in the case of a photoexcited semiconductor [6, 10]. The Hamiltonian $\mathcal{H}_0 + \mathcal{H}_g$ is identical to two independent Jaynes-Cummings models, one for each cavity mode. A key quantity that governs this system is the detuning $\delta \equiv E_g - \hbar\omega$. From the 4 possible electron-hole pairs, two 'bright' $|X_B\rangle$ pairs are coupled to the cavity modes ($|CM\rangle$)and two 'dark' $|X_D\rangle$ pairs are decoupled from the rest. The spectrum of $\mathcal{H}_0 + \mathcal{H}_g$ is shown in fig. 1, for different values of δ in the manifold of $\mathcal{N} = 1$ excitation (see below). The levels have a trivial $(2S+1)^2$ degeneracy of the uncoupled local spins $S = 5/2$. The ground state manifold is mainly photonic in the $\delta > 0$ case (1a), mainly excitonic ($|X_B\rangle$) in the $\delta < 0$ case (1c) and it is a compensated mixture in the $\delta = 0$ case (1c) . The exchange interaction between the fermions and the spin $S = 5/2$ of the Mn impurities reads:

$$\mathcal{H}_J = \sum_{I,f} J_f \vec{S}_I \cdot \vec{S}_f\left(\vec{x}_I\right) \tag{3}$$

where $\vec{S}_f\left(\vec{x}_I\right)$ stands for local spin density of the $f = e, h$ fermion and $\vec{S}_I$ is the Mn spin located in $\vec{x}_I$. The electron spin density reads: $\vec{S}_e\left(\vec{r}_I\right) = \frac{1}{2}|\psi_e(\vec{r}_I)|^2 c^\dagger_\sigma c_{\sigma'} \vec{\tau}_{\sigma,\sigma'}$ and analogously for the holes. The strength of the interaction between the quantum dot fermion and the magnetic impurity depends both on the exchange constant of the material J_f and on the localization degree of the carrier determined by the confinement of the dot, $|\psi_f(\vec{r}_I)|^2$. We consider a hard wall quantum dot, with lateral dimensions $L \simeq 10$ nm and total volume $\Omega_{QD} \simeq 1200$ nm^3. The upper limit for the exchange interaction between a conduction (valence) band electron (hole) and a Mn spin is $j^{max}_e = 8 \times \frac{J_e}{\Omega_{QD}} = -0.1$meV ($j^{max}_h = 8 \times \frac{J_h}{\Omega_{QD}} = +0.5$meV). The exchange coupling can be recasted as $\frac{1}{2}\eta(\vec{x}_I) j^{max}_f \vec{\tau}_f \cdot \vec{S}_I$. For a given dot, $0 < \eta < 1$. Larger exchange interaction ($\eta > 1$) can be obtained in smaller dots.

The last term in the Hamiltonian describes the intra-band U_1 and inter-band U_2 Coulomb interactions between the quantum dot electrons[20]:

$$\mathcal{H}_U = U_1\left(n_{c,\downarrow} n_{c,\uparrow} + \overline{n}_{d,\downarrow}\overline{n}_{d,\uparrow}\right) + U_2 \sum_{\sigma,\sigma'} n_{c,\sigma}\overline{n}_{d,\sigma'}, \tag{4}$$

where $n_{c,\sigma} = c^\dagger_\sigma c_\sigma$ and $\overline{n}_{d,\sigma} = 1 - d^\dagger_\sigma d_\sigma$. When the interaction is included, we have $E_G = \epsilon_c + \epsilon_v - (U_1 - U_2)$.

The band gap in (Cd,Mn)Te is the largest energy scale in the problem. We have verified numerically that two consequences follow. First, the effect of the terms that do not conserve the number of excitons plus photons in $\mathcal{H}_g$ is fully negligible and they can safely be removed from the Hamiltonian. This is known as the rotating wave approximation and permits to decompose the Hilbert space in uncoupled and numerically tractable subspaces with $\mathcal{N}$ excitations. Second, the ground state of the problem, which lies in the $\mathcal{N} = 0$ sub-space, has a extremely small Mn-Mn coupling due to accross-gap Bloembergen-Rowland coupling. Here we consider the Mn-Mn coupling, in presence of $\mathcal{N} > 0$ polaritons and we solve *exactly* the Schrodinger equation $\mathcal{H}|\Phi^{\mathcal{N}}_i\rangle = E^{\mathcal{N}}_i|\Phi^{\mathcal{N}}_n\rangle$.

Results for $\mathcal{N} = 1$. In this manifold the effect of $\mathcal{H}_U$ is trivial. For simplicity, we take $U_1 = U_2 = 0$ in this case. Figure 1 (symbols) shows the excitation spectrum $E_i - E_G$, where E_G is the ground state of the $\mathcal{N} = 1$ manifold and i labels the excited states, for $g = 5$ meV and 3 different detunings, $\delta = -2 \times g$ (fig 1a), $\delta = 0$ (fig 1b) and $\delta = 2g$ (fig 1c). The flat lines correspond to the energy spectrum without exchange coupling. The dispersion of the energy levels, compared with the case without exchange, indicates the strength of the indirect exchange interaction and, as seen in figure 1, is proportional to the excitonic content of the levels. In order to quantify how the impurities are correlated we define a spin-spin correlation:

$$\langle\vec{S}_1\cdot\vec{S}_2\rangle_{\mathcal{N}} = \frac{1}{Z_{\mathcal{N}}}\sum_i \langle\Phi^{\mathcal{N}}_i|\vec{S}_1\cdot\vec{S}_2|\Phi^{\mathcal{N}}_i\rangle e^{-E^{\mathcal{N}}_i/kT} \tag{5}$$

where $Z_{\mathcal{N}}$ is the partition function and the sum runs over the eigenstates Ψ_i of the Hamiltonian with energies E_i

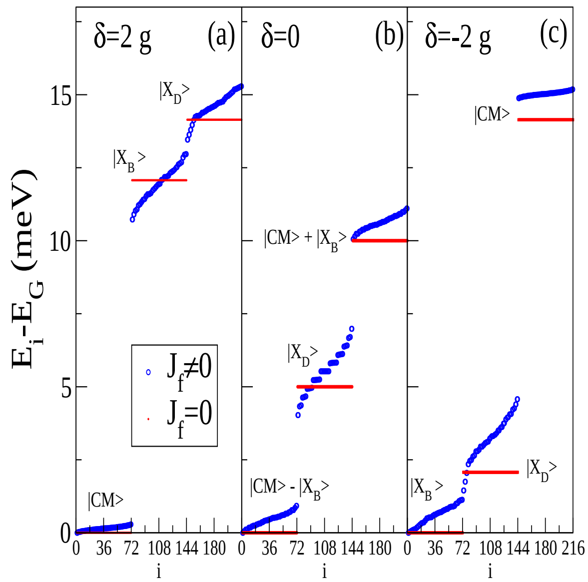

FIG. 1: Spectrum of $\mathcal{H}$ for $g = 5$ meV and different values of δ, S, with (symbols) and without (points) exchange.

with $g = 0$ (fig. 2d) has $2 \times \langle S_z^1 S_z^2 \rangle_{\mathcal{N}} = \langle S_\perp^1 S_\perp^2 \rangle_{\mathcal{N}}$. This proves that $\mathcal{H}_g$ is responsible for the anisotropy of the spin correlations seen in figs. 2b and 2c.

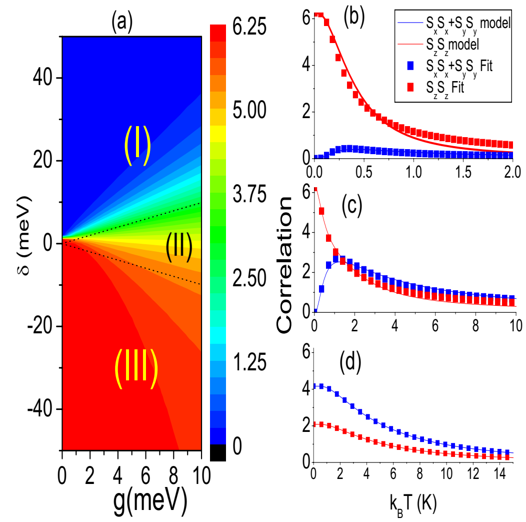

FIG. 2: Left: Contour plot for $\vec{S}^1 \cdot \vec{S}^2$ as a function of g and δ for $k_BT = 1$K. Right: Correlation functions both for $\mathcal{H}$ (lines) and $\mathcal{H}_{eff}$ (symbols)

in the $\mathcal{N}$ manifold. This correlation function corresponds to a density matrix in which the degrees of freedom are in equilibrium inside the manifold with $\mathcal{N}$ polaritons. In figure 2a we plot $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle_{\mathcal{N}=1}$ as a function of δ and g at $k_bT = 1$K and $U_1 = U_2 = 0$. Results with finite values of U_i can be obtained exactly by replacing $\delta(U_1 = U_2 = 0)$ by $\delta(U_1 = U_2 = 0) - (U_2 - U_1)$. The phase diagram has 3 different regions. In region I ($\delta > g > 0$), the polariton is mainly photonic but, because of the light-matter coupling g, it has a small excitonic component that correlates the spins. In this region the spin correlation vanishes identically if $g = 0$. This is the optical exchange interaction region. In region (II) ($|\delta| < g$) the polaritons have a large content of both exciton and photon that is properly captured by our non-perturbative approach. Region (III) ($\delta > g > 0$) the polariton is mainly excitonic and the spin correlation comes from carrier mediated exchange interaction region. Since the weight of excitonic part in the wave function is larger than in regions (I) and (II) the interactions are consequently stronger. The segment $g = 0, \delta < 0$ in fig. 2a (see also 2d) corresponds to a bare quantum dot, totally decoupled from the cavity, and occupied by 1 exciton. In this case a significant ($S^2/3$) correlation survives at 15 Kelvin (fig. 2c). Figures 2a and 2b support our claim that confinement of both the cavity mode and the excitons enhances optical exchange interaction to the point that *a single photon* correlates 2 distant spins at $k_BT = 1$K.

In figure 2b,2c and 2d we plot $\langle S_z^1 S_z^2 \rangle_{\mathcal{N}}$ and $\langle S_\perp^1 S_\perp^2 \rangle_{\mathcal{N}}$ as a function of temperature for region (I) (fig. 2b), region (III) with finite g (2c) and the purely excitonic case ($g = 0$, 2d). Figures 2b and 2c describe the same system than figures 1a and 1b respectively. Only the case

A simple estimate of ordering temperatures in dots with many spins can be obtained if the low energy sector of our model is described with an anistropic Heisenberg Hamiltonian $\mathcal{H}_{eff} = -J_x \left(S_x^1 S_x^2 + S_y^1 S_y^2 \right) - J_z S_1^z S_2^z$. We find J_x and J_z by fitting the static spin correlation function of eq.(5) to that of the Heisenberg model (fig. 2(b,c,d)). Remarkably, it is always possible to find J_x and J_z such that correlations functions are similar to a few percent in a wide temperature range. The outcome of the procedure is stable with respect to small variations of parameters in the problem. We obtain J_x =15mK and J_z =0.13K for $\delta = 2g = 10meV$ (fig 2b), J_x =0.35K and J_z =0.58K for $\delta = -2g = -10meV$ (fig 2c), $J_x = J_z$ 0.6K for $g = 0$, $\delta < 0$ (fig 2d). Using the mean field result $k_BT_c = \frac{S(S+1)}{3} zJ$, where z is the typical number of Mn coupled to a given one, we obtain $k_BT_c \simeq z \times 2$K for the case of fig. 2d, that corresponds to exciton mediated coupling. The exciton mediated spontanenous polarization in CdTeMn quantum dots surviving at 120 Kelvin recently reported [13], could be justified if we take $z \simeq 60$ wich is not unreasonable [9]. The above results are obtained for $\eta = 1$. In figure 3d we plot the correlation function for the same system of figs. 1a, 1b and 1c as a function of η at 1 Kelvin. For $\eta < 1$ ($\eta > 1$) these curves reflect how the spin spin correlations are degraded (enhanced) as the fermion's wave function is less (more) peaked on the Mn positions. The region $\eta > 1$ corresponds to dots smaller than the $\eta = 1$ reference.

Results for $\mathcal{N} > 1$. Optical exchange interaction in

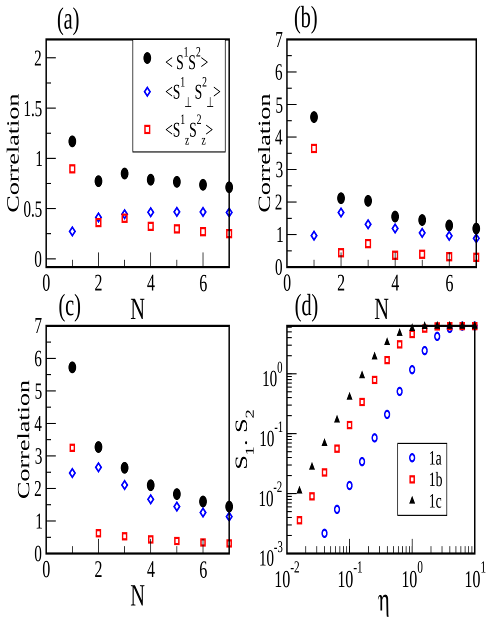

FIG. 3: (a), (b), (c): spin correlation as a functions of N for the same parameters than fig. 1a,1b and 1c respectively. (d): spin correlation function as a function of η.

bulk is proportional to the square of the electromagnetic density. In the Jaynes-Cummings model the light matter coupling is renormalized like $g_{\mathcal{N}} = \mathcal{N} g$ and figure 2a shows how the spin correlation is an increasing function of g in regions (I) and (III). Hence, it might seem that increasing $\mathcal{N}$ should increase the effective coupling. In contrast, double occupancy of the fermion levels in the dot blocks the possibility of spin exchange with the impurities and dramatically reduces the effective interaction [9]. The relative importance of these competing factors is tuned as δ changes. Interestingly, intra-band Coulomb repulsion reduces the double fermion occupancy and favors the ferromagnetic exchange. Figs. 3a,3b and 3c display the spin correlation function for $k_B T = 1$ K as a function of $\mathcal{N}$ for the same cavity-dot systems of figures 1a, 1b and 1c respectively, with U_1=20 meV and $U_2 = 10$ meV. In the three cases the total correlation decreases as the number of cavity exciation increases, reflecting that Pauli blocking overcomes the enhancement of Rabi energy. The dominant axis of the spin correlation undergoes a crossover from off plane (z) to in plane (xy) that reflects how the change in the relative weight of the bright and dark excitons as the Rabi coupling increases. This crossover opens the door to optical tuning of the easy axis in the ferromagnetic phase.

Discussion and conclusions. We have studied the indirect exchange interaction between two spins in a cavity-dot system with $\mathcal{N}$ exciton polaritons. In practice the Fock cavity states could be prepared by pumping the cavity with the adequate laser[21], although some electronic injection method could be devised[22]. The detection of the resulting spin correlations could be done by spin resolved photoluminescence detection [13] or by pump and probe [8]. The exact diagonalization of our model permits to study the evolution of the indirect exchange interaction as the cavity mode energy crosses over the band gap, going from the limit of 'optical RKKY' [6, 10] to conventional carrier mediated interaction. Attending to the number of photons involved, the cavity-dot system provides a huge enhancement of the optical exchange interaction compared with bulk systems [6, 10], since a single photon yields a sizable indirect exchange interaction that survives at 1 kelvin. This makes intense lasers unnecessary in the implementation of the optical exchange interaction.

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